

**Determination of Interfacial Tension of Immiscible Two-Phase, Oil-Water
System Subjected to Polymeric Drag Additives**

by

Nurul Sakinah binti Rosli
15618

Dissertation submitted in partial fulfilment of
the requirements for the
Bachelor of Engineering (Hons)
(Chemical Engineering)

SEPTEMBER 2015

Universiti Teknologi PETRONAS,
32610, Bandar Seri Iskandar,
Perak Darul Ridzuan

CERTIFICATION OF APPROVAL

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A project dissertation submitted to the
Chemical Engineering Programme
UniversitiTeknologi PETRONAS
in partial fulfillment of the requirement for the
BACHELOR OF ENGINEERING (Hons)
(CHEMICAL ENGINEERING)

Approved by,

(Mr MohdZamri Abdullah)

UNIVERSITI TEKNOLOGI PETRONAS

BANDAR SERI ISKANDAR, PERAK

September 2015

CERTIFICATION OF ORIGINALITY

This is to certify that I am responsible for the work submitted in this project, that the original work is my own except as specified in the references and acknowledgements, and that the original work contained herein have not been undertaken or done by unspecified sources or persons.

NURUL SAKINAH BINTI ROSLI

ABSTRACT

In this paper, the determination of interfacial tension of immiscible two-phase oil-water system subjected to polymeric drag additives was investigated. Polymeric drag reducing additives (PDRA) have been discovered in the late 1940s, where its first large-scale commercial utilization was implemented in the Trans-Alaskan Pipeline System (TAPS) three decades after its discovery by Toms. The ability of polymer solution to modify the viscosity of the flowing medium translates into its assistance of altering the flow properties during the transportation of fluids. During the process, PDRA promoted the bubbly and dispersed flow into stratified regime, where oil-water is separated via a pronounced interfacial boundary. In this condition, the nature of the interfacial tension is unknown due to the changes caused by the dilution of PDRA into the flowing medium. This research presents the study on the rheological properties of the partially hydrolysed polyacrylamide (PHPA) solution that acted as PDRA. The experimental work consists of viscometry and the pendant drop analysis. Three different concentration; 10 ppm, 50 ppm and 70 ppm from a 1000 ppm master solution of PHPA were tested at elevated temperature to study its rheological properties as well as the interfacial tension of the oil-water phase. From the results, it is clear that higher concentration of PHPA leads to higher viscosity. All solutions that were investigated in this study exhibited “shear thinning”, and is proven as the n value calculated is less than 1. From the results, the PHPA solutions investigated behaved in a non-Newtonian manner that abide the Power Law. The addition of all the PHPA solution had decreased the interfacial tension of the oil-water system.

ACKNOWLEDGEMENT

First and foremost, I would like to thank and appreciate my parents, Mr Rosli bin Abdullah and Mrs Poziah binti Yusuf for their material and spiritual support in all aspects of my life.

Besides my parents, I would like to express my sincere gratitude to my supervisor, Mr M. Zamri Abdullah for the immense knowledge and thoughtful guidance especially during conducting the experiment and writing process. I am sincerely grateful to him for sharing his illuminating views related to the project.

Not to forget, deepest appreciation to Mr. Hazri from Block 21 and Mr. Awais from Block P for facilitating me on using the equipment at the department. Their willingness to spend time to assist in every difficulty that I faced is much appreciated.

Last but not least, I would like to express my heartfelt gratefulness to my friends who gave me continuous support in completing this thesis.

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CHAPTER 1

INTRODUCTION

1.1 Background of Study

The phenomenon of drag reduction by polymer additives has almost exclusively been studied since the pioneering work of Toms in the 1940s. Ever since Toms finding suggested on the use of polymer additive to enhance the flow of crude oil in the pipeline, the research on drag-reducing additives (DRA) has increased ever since. After almost 3 decades, the first commercial use of a polymeric drag-reducing additive to increase the flow rate in a crude oil pipeline began during 1979 in the Trans Alaska Pipeline System (TAPS). It was one of the most impressive successes in polymer applications for drag reduction in advanced production systems. The ability of polymer solution to modify the viscosity of the medium translates into its assistance of altering the flow properties during the transportation of fluids. In a multiphase system consisting of immiscible liquids, polymeric drag reducing agent (PDRA) could alter the interface between the phases, particularly during stratified flow. This change would affect the shape of the curvature experienced during the flow to be either flat, concave upward or concave downward, which further decodes to the modification of pressure drop during the flow.

With the application of drag reducing agents (DRA), negative consequences of pipeline pressure losses can be avoided. Since the impressive successes in drag reducing agents application in TAPS, DRA have been conventionally used in the oil and gas industries (Al-Sarkhi, 2010). Due to its practicability, PDRA could reduce the usage of pumps and provide better efficiency in flow of liquid in pipeline.

DRA can be split into three groups: polymers, surfactants and fibers. There are no general guidelines for the selection of a DRA for a given multiphase flow application. The most significant requirement is that the DRA is soluble in the liquid that is being tested (Mowla & Naderi, 2006). For this experiment, partially hydrolysed polyacrylamide (PHPA) is used where it has a variety of application in the oil and gas industry such as in improving the production of oil, reducing the friction, as a fluid loss control and for lubrication. High molecular weight polymer helps in reducing the Reynolds shear stress and varying velocity (Al-Sarkhi, 2010).

Studies involving Drag Reducing Polymer (DRP) in two-phase oil–water flow are not only very limited, but also started just recently in less than a decade ago (Abubakar, Al-Wahaibi, Al-Hashmi, et al., 2015). Unlike the use of DRPs in single phase and two-phase gas–liquid flows which have received exhaustive attentions since its discovery, very little experimental data on the use of DRPs in oil–water flow are available in the literatures. Most of the studies are about pressure drop, drag reduction and the efficiency of the PDRA and not much considered the impact of PDRA towards the change of interfacial curvature of the flowing fluids in pipe that may affect the overall pressure drop. As PDRA is added into the flowing medium and dissolved accordingly, the change of the in-situ viscosity is predicted to influence the shape of the curvature due to the wettability towards the wall and two fluids interfaces. This study, therefore will investigate the impact of partially hydrolyzed polyacrylamide (PHPA) added into the liquid medium (solvent) by looking into the change of in-situ viscosity and its influence to the interfacial curvature in two-phase liquid system.

1.2 Problem Statement

PDRA has been widely applied in the pipeline system to reduce the drag forces between the oil and the pipeline wall in order to increase the oil throughput. When PDRA is injected into a stratified flow of oil-water phase, the shape of the curvature of the multiphase changes accordingly. This study will investigate the effects of DRP towards the interface that also influence the alteration of the interface's curvature shape.

1.3 Objective

Following are the specific objectives of this work:

- i. To investigate the rheological properties of diluted concentration of PHPA through shear analysis at various temperatures.
- ii. To determine the effect of dilute PHPA to the interfacial tension in the oil-water, two phase condition.

1.4 Scope of Study

The PHPA acts as a polymeric drag reducing agent (PDRA) in the pipeline system which transports oil. For this study, the rheological properties of PHPA at different concentration will be analyzed by using rheometer. The PHPA powder will be mixed with water to produce a solution at different concentration. The experiment will be conducted using different concentrations of PHPA at a varying temperature. Since many studies has focused on how the DRP affect the pressure drop inside the pipeline, this study will be focused on the shear analysis and the effect of using PHPA to the interfacial surface tension in oil-water phase.

CHAPTER 2

LITERATURE REVIEW & THEORY

2.1 Polymeric Drag Reducing Agent

According to Karami and Mowla (2013), primary studies on drag reduction were conducted by Toms about forty years ago. Toms observed that a substantial reduction of the frictional pressure gradient could be achieved by the addition of 10 ppm by weight of polymethylmethacrylate to turbulent monochlorobenzene flowing down the pipe (Mowla&Naderi, 2004). With the addition of very small amounts of polymers in liquid, it can suppress turbulent flow, or at least reduce turbulent losses to a great degree.

The evolvement of polymeric drag reducing agent (PDRA) is from the complication that is caused by transporting the multiphase mixture from hundreds of kilometres to the separating tank. In order to overcome the difficulty in separating the phases, the PDRA needs to be introduced. Abubakar et al. (2014) stated that polymer have been found to offer frictional drag reduction of turbulent flow which leads to savings in energy consumption and economic relieve by eliminating the need to install pumping stations. The authors also stated that the high molecular weight polymers assist to suppress the formation of turbulent bursts in the buffer region, and in turn restrain the formation and propagation of turbulent eddies (Figure 1). In addition, PDRA does not only help in occurrence of pressure drop, it also contributes in maintaining the stratified flow which would help in the separation of oil-water phase (Al-Sarkhi, 2010).

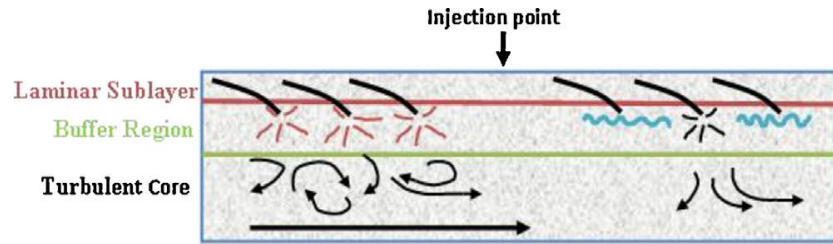


FIGURE 2.1 Illustration of pipeline turbulent flow regions (Abubakar, Al-Wahaibi, Al-Wahaibi, Al-Hashmi, & Al-Ajmi, 2014)

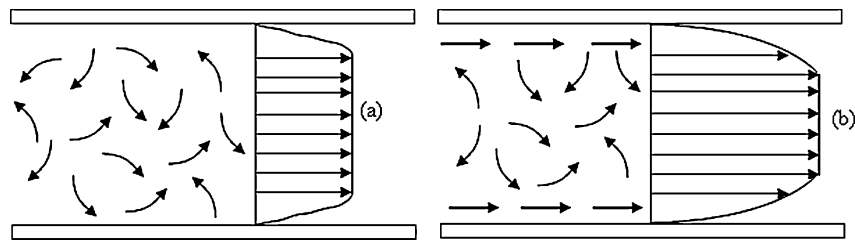


FIGURE 2.2 Velocity profiles of the turbulent flow of (a) a pure liquid and (b) a liquid that contains a polymer additive (Abubakar et al., 2014)

Despite the great number of studies regarding drag reducing polymers, the underlying mechanisms of drag reduction are yet to be clearly defined. Lumley (1969) proposed a mechanism for the dynamics of polymer by suggesting that the elastic properties of polymers and elongation of coiled polymer molecules increases the thickness of viscous sublayer (Karami & Mowla, 2013). This prevents the hydraulic energy provided by the pumps in creating a chaotic and random motion. Instead, the energy is more directed in moving the fluid down the pipeline. For that reason, polymer can produce drag reduction up to 80% which makes them the most studied and highly-employed drag reducing agent in the industries (Abubakar et al., 2014).

Studying on the effect of different concentrations of PDRA in a slug flow of air and crude oil, Mowla and Naderi (2006) reported that the optimum concentration needed to give the highest drag reduction is 18 ppm. However, the efficiency of PDRA are also influenced by other factors than the concentration, such as the size and the type of the pipe used as well. Khadom and Abdul-Hadi (2014) further reported that the drag reduction percent is increased with the increase in velocity and concentration of additive. Higher drag reduction resulted from the increasing concentration of additive

was also seen by Pereira, Andrade, and Soares (2013) using polyethylene oxide, polyacrylamide, and xanthan gum.

Abubakar et al. (2015) conducted an experiment by adding 40 ppm DRP and observed that the addition of DRP changed the stratified wavy flow to stratified flow pattern which then reduced the amplitude of the wavy formation at the oil-water interface. From the experiment, it can be seen that the drag reductions by DRP also depend on the flow structures of the oil–water flow.

This particular study will utilize partially hydrolysed polyacrylamide (PHPA) as the PDR since it is commonly used as PDRA due to its flexible molecule and highly soluble in water (Khadom & Abdul-Hadi, 2014). It is in white dry-solid form with an average molecular weight of 5×10^6 g/mol.

2.2 Stratified Flow

Stratified flow is defined as two fluids flow in separate layers according to their different densities, where the heavier phase will tend to flow near the bottom of the conduits. liquid-liquid mixture tends to separate and flow at different velocities due to density difference. As oil-water mixtures are difficult to separate at the end of pipeline, conserving the stratified pattern for a wider range of conditions would facilitate the separation of the oil and water (Al-Wahaibi, Smith, & Angeli, 2007). Stratified flow is easier to occur when the velocity is low but as the velocity increases, the flow of the multiphase liquid changes to non-stratified and finally to dispersed flow. Angeli and Hewitt (2000) concluded that the variables influencing the flow patterns are density difference, oil viscosity and also the wetting properties of the wall.

According to Xu (2007), each layer will be defined as either water or oil continuous based on the inversion point. In liquid-liquid system with small density difference or in reduced gravity with high density difference, Brauner, Moalem Maron, and Rovinsky (1998) proposed that the surface phenomena may dominate the flow and create a curved interface. Depending on the physical properties of the fluids, solid-fluid wettability, the geometrical dimensions and the fluids hold-up, the free interface

may attain a plane or curved configuration. Generally, when surface effects are significant, the interface configuration tends to attain a convex or concave configuration depending on the relative wettability properties of the two fluids with the wall surface. On the other hand, the interface approaches a plane configuration when the gravity is dominant. A two-fluid model for analyzing oil-water stratified flow with curved interfaces are as below.

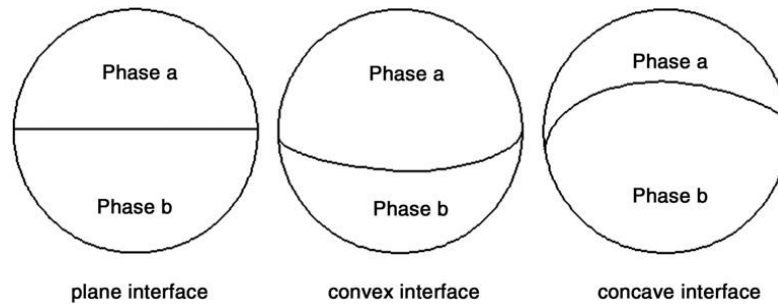


FIGURE 2.3 Schematic description of two phase stratified flow with curved interfaces (Xu, 2007)

Abubakar et al. (2015) investigated the relationship between drag reduction and slip velocity ratio of oil-water flow in a horizontal acrylic pipe. The experiment conducted showed that the interface of oil–water exhibited a concave shape. When the oil and water are in separated flows, the oil will maximize its contact area with the pipe as oil naturally has a preferential ability of wetting acrylic pipe. Therefore, the frictional drag by the wall will have more effect on the oil and thereby slowing down the oil phase as compared with the water phase. Another reason for occurrence is the fact that the viscosity of the oil is more than that of water and hence, the oil travelled slower than water.

Meanwhile, Al-Wahaibi et al. (2007) stated that in stratified flow the interface is either smooth or wavy with small and large amplitude waves. Using a two-fluid model it was found that both interfacial and water wall shear stresses decrease when polymer is present in the water phase. The interfacial stress is probably reduced because the interface becomes smoother, while the reduction in the water wall stress could be a combination of decreased water velocity and turbulence.

2.3 Interfacial Tension of Oil-Water Phase

Interfacial tension (γ) is described as the force acting on the interface resulted from the influence of the aligned molecules at each of the phase's interface (Cao & Li, 2002). It is the measure of how much energy is required to make a unit area of interface between two immiscible liquids. The interface can also be between a gas and a liquid however, this tension is typically termed as surface tension (σ).

For two immiscible fluids, the interfacial tension arises from the dissimilarity of the intermolecular forces between the molecules in the phases (Isehunwa & Olubukolu, 2012). The interfacial tension of immiscible liquids is a significant physical property that is useful in determining the behaviour of liquids in such diverse areas such as dispersions, emulsions, and enhanced oil recovery processes (Kim & Burgess, 2001). Hyde, Phan, and Ingram (2014) stated that the liquid-liquid interfacial tension is one of the main physical parameter that effects the multiphase system in a wide range of processes, which include separation and emulsification that are widely used in the chemical industry.

Interfacial tension of heavy crudes depend on temperature, salt concentration and viscosity (Isehunwa and Olubukola, 2012). It was observed that interfacial tension increases with increasing temperature in light oil-brine systems but decrease with increasing temperature in heavy crude-brine systems. Early researches discovered that by reducing the interfacial tension between crude oil and connate water to ultralow, or $<10^{-2}$ mN/m, for example, using surfactants, can help to recover the oil droplets trapped in porous locks due to capillary action (Pei, Yu, Hu, & Cui, 2014). Mosayeb and Abedini (2012) cited that the interfacial tension plays a fundamental role in conventional and enhanced oil recovery (EOR) methods. The use of surfactants in enhancing oil recovery has generally based on the reduction of the interfacial tension between the crude oil and the flooding phase which can be performed by reducing the capillary forces to improve the microscopic displacement efficiency. This method is also known as tertiary oil recovery technique or surfactant flooding. The displacement of particle between the liquid phases is correlated to the oil-water interfacial tension and contact angle and is illustrated in Figure 4.

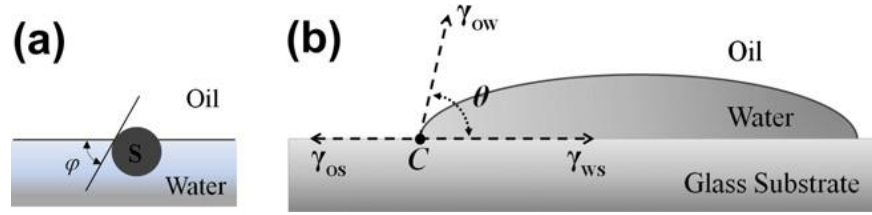


FIGURE 2.4 Definition of the contact angle that a particle assumes at the oil-water interface b) energetic configuration of a liquid drop on a glass surrounded by oil (Pichot, Spyropoulos, & Norton, 2012)

Increasing the surfactant concentration will usually decreases the oil/water interfacial tension (Pei et al, 2014). This was also confirmed by a study by Pichot et al. (2012) on the effect of hydrophilic silica particles in the presence of surfactant on the interfacial tension of oil and water. They reported that increasing the surfactant concentration had decreases the interfacial tension of the oil/water system (Figure 5). At low surfactant concentration the interfacial tension is effected by the silica particles which increases the interfacial tension , while at high surfactant concentration, the interfacial tension is unaffected by the silica particles and are only dictated by the surfactant concentration.

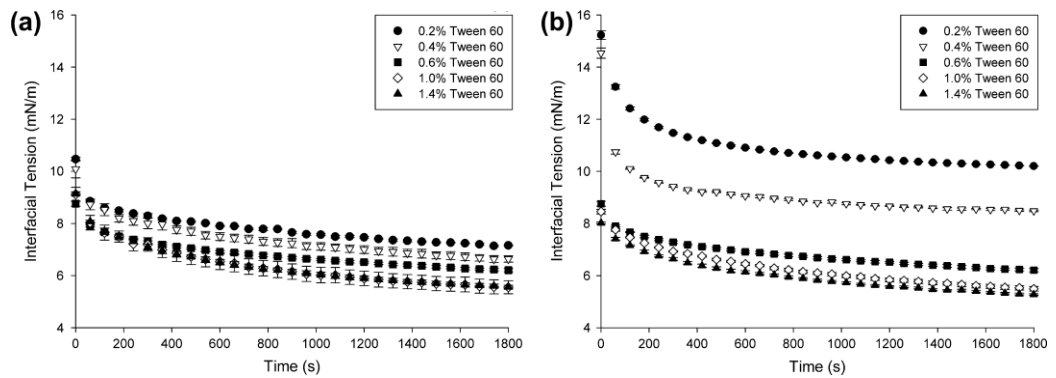


FIGURE 2.5 Interfacial tension versus time of systems containing water-oil-Tween 60 in the absence (a) or presence (b) of hydrophilic silica particles (Pichot et al., 2012)

The authors further explained that surfactants replace the molecules of water and oil at the interface. The interactions between the surfactant and water molecules at one side and oil molecules at the other side are much stronger than the original oil/water interactions, which then significantly reduce the interfacial tension.

From an experiment conducted by Abubakar, Al-Wahaibi, Al-Wahaibi, et al. (2015) on the effect of low interfacial tension on flow patterns, it shows that low interfacial tension enhances the phase mixture as the mixing or the emulsion of one phase in another phase is controlled by the interfacial tension between the two phases. There was no significant difference on flow patterns or pressure drop between the two polymer concentrations used. However, the higher polymer concentration appears to damp more the interfacial waves.

The following formula is used to determine the interfacial tension increment per unit fractional change in the interfacial area:

$$\varepsilon = \frac{d\gamma}{d \ln A} = |\varepsilon| \exp(i\theta) \quad (1)$$

Where,

ε : Dilatational modulus

γ : Interfacial tension

θ : Phase angle

A : Area of interface

Kim and Burgess (2001) developed an equation which uses a nonlinear regression in order to get the interfacial tension between the miscible organic fluids over its composition range. Through experimentations using five types of oil mixtures, the authors concluded that the interfacial tension is affected by the volume fraction and the interfacial tension difference of the two pure oils. The equation that was developed is able to predict the interfacial tension of miscible multiphase mixtures. Interfacial tension of oil-water phase could be determined by the following equation:

$$\gamma = (\gamma_1 - \gamma_2) \exp(\alpha V^{0.7}) + \gamma_2 \quad (2)$$

Where,

γ : Interfacial tension

$\gamma_1 - \gamma_2$: Interfacial tensions of pure oils/water where $\gamma_1 > \gamma_2$

α : Exponential coefficient

V : Volume fraction of the oil mixture

Gülseren and Corredig (2014) studied the interactions between commercial pectins and polyglycerolpolyricinoleate (PGPR) at the oil-water interface and found that the usage of sugar beet pectin (SBP) has caused a declination in the interfacial

tension synergistically with PGPR while High methoxyl pectin (HMP) affects the interfacial activity where it causes a difference in the viscoelastic properties of the interface.

Cai, Yang, and Guo (1996) measured the interfacial tension of 10 normal alkane/water with brine and hydrocarbon mixtures/water with brine systems in a study where the effects of temperature, pressure, and salt content were assessed. It was found that the behavior of the interfacial tension was sensitive to temperature as it decreases with increasing temperature.

2.4 The Pendant Drop Method

There are numerous techniques that were proposed to measure the interfacial tension which are the Wilhelmy plate, maximum bubble pressure, spinning drop, Du Nouy Ring, capillary rise and the pendant drop (Berry, Neeson, Dagastine, Chan, & Tabor, 2015).

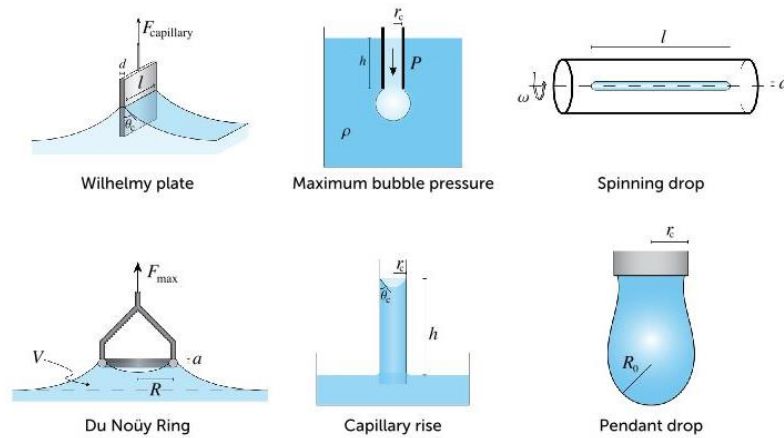


FIGURE 2.6 Schematics of various experimental techniques used to determine interfacial tension (Berry et al., 2015).

The pendant drop method which employs Axisymmetric Drop Shape Analysis (ADSA) has remained the most practical method for tensiometric analysis of liquid-liquid systems (Hyde et al., 2014). Berry et al. (2015) further stated that by easily suspending the fluid droplet from a needle, the pendant drop tensiometry is arguably the simplest, most potent and most versatile among these methods. ADSA methods are applicable to pendant and sessile drops in a very wide range of surface tensions,

for any fluid-liquid system that can be represented by the Laplace equation of capillarity (Río & Neumann, 1997). This method is an extensively used drop shape technique for surface tension and contact angle measurement that uses specialized analysis software with high resolution images to match experimental drop profiles with solutions to the Young-Laplace equation of capillary

The pendant drop method uses the concept of axis symmetric fluid bodies which are analysed in a vertical direction as it is affected by gravitational forces that affects the surface curvature. In 1980, Boucher has figured eight ways to get the interfacial equation arrangement. The equations are further described by the Young-Laplace equation and is expressed by three ordinary differential equations:

$$\frac{d\theta}{dS} + \frac{\sin \theta}{X} = 2\lambda(H - Y) \quad (3)$$

$$\frac{dX}{dS} = \cos \theta \quad (4)$$

$$\frac{dY}{dS} = \sin \theta \quad (5)$$

$$a = \sqrt{g\Delta\rho / \gamma} \quad (6)$$

$$Bo = \frac{\Delta p g R^2}{\gamma} \quad (7)$$

Where:

S: Distance along the drop surface and the meridian angle

θ : Angle from the horizontal plane

λ : Type of drop

H : Shape factor

X : Radial coordinate

Y : Vertical coordinate

a : Capillary length to normalized the coordinates

Bo : Characterization of the deformation of the interfacial tension

R : Characteristics of length in the system

γ : Interfacial tension

Saad, Policova, and Neumann (2011) mentioned that pendant drop method has apparent advantages of simplicity and flexibility, and also high accuracy. Other than that, Woodward (n.d.) mentioned that solid surfaces of the apparatus involved in pendant drop need not have any special cleanliness because their wettability does not affect the result. This is a significant advantage over such techniques as the Wilhelmy plate where cleanliness is essential.

Generally, the shape of the pendant drop depends on the balance between gravity and surface tension as reflected mathematically in the Laplace equation of capillarity. The surface tension can be determined from an analysis of the shape of the pendant drop when the gravitational and surface tension effects are comparable. The surface tension tends to round the drop, whereas gravity deforms it and tends to elongate a pendant drop. The shape of the drop will tend to become close to spherical whenever the surface tension effect is much higher than the gravitational effect.

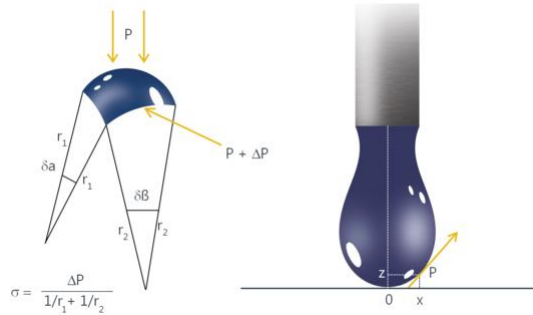


FIGURE 2.7 The Pendant Drop Method

According to Woodward (n.d.), the principal assumptions of drop shape analysis are the drop is symmetrical from the vertical axis which makes it insignificant from where the drop is viewed. Also, the viscosity and inertia are not affecting the shape of the drop as it is not in motion. This means that only the interfacial tension and gravity forces are shaping the drop. The relationship between interfacial pressure and these radii of curvature is called the Young-Laplace equation of capillarity:

$$\Delta P = \sigma \left(\frac{1}{r_1} + \frac{1}{r_2} \right) \quad (8)$$

Where,

ΔP = Pressure drop

σ = interfacial tension

r = radius of curvature

2.5 Viscosity

Mobility reduction or viscosity behavior of partially hydrolyzed polyacrylamide polyelectrolyte solutions plays an important role in enhanced oil recovery. A dilute aqueous solution of partially hydrolyzed polyacrylamide is used as a pushing fluid in the injection wells to sweep oil in the reservoir into the production well (Zeynali, Rabii, & Baharvand, 2004). According to Gao (2013), polymer increases the viscosity of injected water, reduces water mobility, and therefore achieves a more stable displacement.

CHAPTER 3

METHODOLOGY

3.1. Experimental Activities

The experimental work in this project can be divided into three parts. The experimental works are as follows:

3.1.1. Preparation of PHPA solutions

The polyacrylamide powder was added to distilled water and stirred for 24 hours to ensure good mixing to prepare the a 1000ppm master solution. Then, the diluted solution of 10ppm, 20ppm and 50ppm was prepared from the master solution. The following explains the procedure to prepare the mixture of water and the drag reducing agent that is used for the experiments.

Preparation of 1000 ppm master solution

1. A beaker with 1000mL of distilled water is prepared.
2. 1.00 gram (g) of polyacrylamide powder is mixed with 1000 ml of distilled water.
3. A 3-bladed propeller stirrer of 2 inch diameter is placed inside the beaker and the stirrer is set at minimum speed of 50 RPM. The solution is stirred for 2 hours.
4. The beaker is covered with a plastic sheet.
5. The mixture is left for 24 hours for hydration where the polymers are broken down into monomers.

Preparation of diluted PHPA solution

1. Calculate the exact amount of water needed to dilute the 1000 ppm master solution to 10 ppm, 20 ppm and 50 ppm using $C_1 V_1 = C_2 V_2$.
2. For 10 ppm, 5mL of the master solution is diluted with 500 mL distilled water to prepare 10 ppm solution. Below is the volume of master solution needed to prepare the diluted version with 500 ml of distilled water.

TABLE 3.1 Volume of master solution required to prepare diluted solution

Concentration of the diluted solution	Volume of master solution
10 ppm	5 mL
20 ppm	10 mL
50 ppm	25 mL

3. A 3-bladed propeller stirrer of 2 inch diameter is placed inside the beaker and the stirrer is set at minimum speed of 50 RPM. The solution is stirred for 2 hours.
4. The beaker is covered with a plastic sheet.
5. The mixture is left for 24 hours for hydration.

3.1.2. Rheology Study of the PHPA Solution

The experiment was conducted to study the effect of shear rates on polymer viscosity. The diluted PHPA solution will be tested using Bohlin C-VOR Rheometer at different temperature of 25°C, 40°C and 50°C with constant shear rate from 0.001 to 10 000s⁻¹. During the experiment, strain was forced by a motor and the generated torque was detected by transducer. Each of the PHPA solution behaviour will be studied under the effect of shearing and temperature to understand how it can affect the interfacial tension of the oil-water phase in the next experiment. The cone and plate spindle with 1°/40mm spindle (CP1/40) was used as the measuring system. This is in many instances the ideal measuring system.

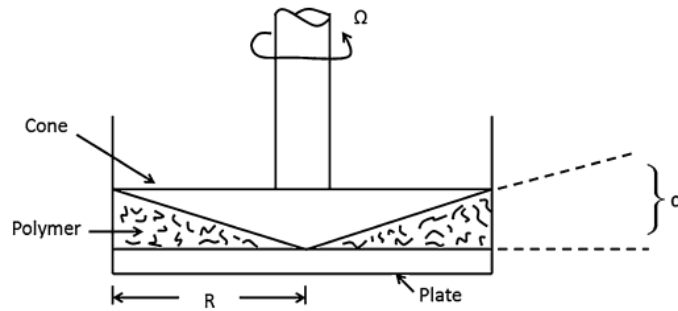


FIGURE 3.1 Schematic of cone-plate rotational viscometer(Stevens, 1999)

The larger the cone angle the more the shear rate across the gap starts to vary, therefore cone angle with 1° is chosen. The following explains the procedure to prepare the PHPA solution to be tested for viscometry experiment in order to study the viscosity of the solution:

1. 1000 ppm solution of PHPA is prepared.
2. The rheometer, heater and air compressor is turned on.
3. The range of shear stress and strain data in the simulation is determined.
4. The shear rate is set as constant which starts from 0 to 10000 1/s.
5. The temperature is set to 25°C, 40°C and 50°C for each tests.
6. The 1/40 spindle is attached with the rheometer and zero the equipment to adjusting the gap size.
7. The sample is placed on the spindle and press start option.
8. The sample is trimmed from the spindle and press the start option again to continue the test.
9. Each test is repeated for 3 times to ensure reproducibility of results.
10. The experiment is repeated by using 10, 20 and 50 ppm of PHPA diluted solution.

3.1.3. Determination of Interfacial Tension of the Oil-Water Phase

Each of the diluted PHPA concentration solution will go through pendant drop method test immersed in diesel with different temperature of 25°C, 40°C and 50°C using the Ramé-hart Model 260 (p/n 260-F4) Goniometer.



FIGURE 3.2 Ramé-hart Model 260 (p/n 260-F4) Goniometer

The microsyringe assembly which contains the PHPA solution helps in exerting the pressure to produce the liquid drop at the needle tip is placed in the environmental chamber which contains diesel oil. The scale of video image of the drop in greyscale mode is measured to get the actual dimension of the drop. The interfacial tension is then calculated using the following equation

$$\gamma = \frac{\Delta\rho g R_o^2}{\beta} \quad (9)$$

γ = interfacial tension

$\Delta\rho$ = difference in mass density

g = gravity constant

R_o = radius of curvature at the drop apex

β = shape factor

Specifications and some physical and chemical properties of the diesel used are listed in Table 2.

TABLE 3.2 Specifications and properties of diesel used as model

Character	Diesel
Density @ 15°C (g/cm ³)	0.830
Viscosity at 40°C (mm ² /s)	3.05

TABLE 3.3 Properties of the polymer solutions

Concentration of PHPA	10ppm	20ppm	50ppm	1000ppm
Physical form	Clear	Clear	Clear	Clear, viscous
Density (g/ml)	0.9972	0.9972	0.9972	0.9972

The following procedure explains the procedure to prepare the PHPA solution to be tested for the interfacial tension studies in order to study the viscosity of the solution:

1. PHPA solution at 10 ppm, 20 ppm and 50 ppm solution of PHPA is prepared.
2. The apparatus is set up.
3. The diesel is poured into the environmental chamber.
4. The PHPA solution is inserted in the microsyringeassy and the needle is placed on the microsyringe assy.
5. The camera and the light is set with light intensity ranges from 30 – 40.
6. Some pressure is exerted to the microsyringeassy to make the shape of the PHPA solution to be like a ‘pear shape’ of a pendant drop.
7. Adjust the sharpness of the image to measure the interfacial tension of the diesel and PHPA solution phase.
8. The temperature of the environmental chamber is set at 25°C, 40°C and 50°C in order to heat the diesel oil.
9. The measurement of the interfacial tension is taken by placing the axisymmetric line on the drop image.
10. The data and calculation is taken from the software.
11. Each test is repeated for 3 times for all three different temperatures at different PHPA solution concentration.

3.2 Gantt chart& Key Milestone

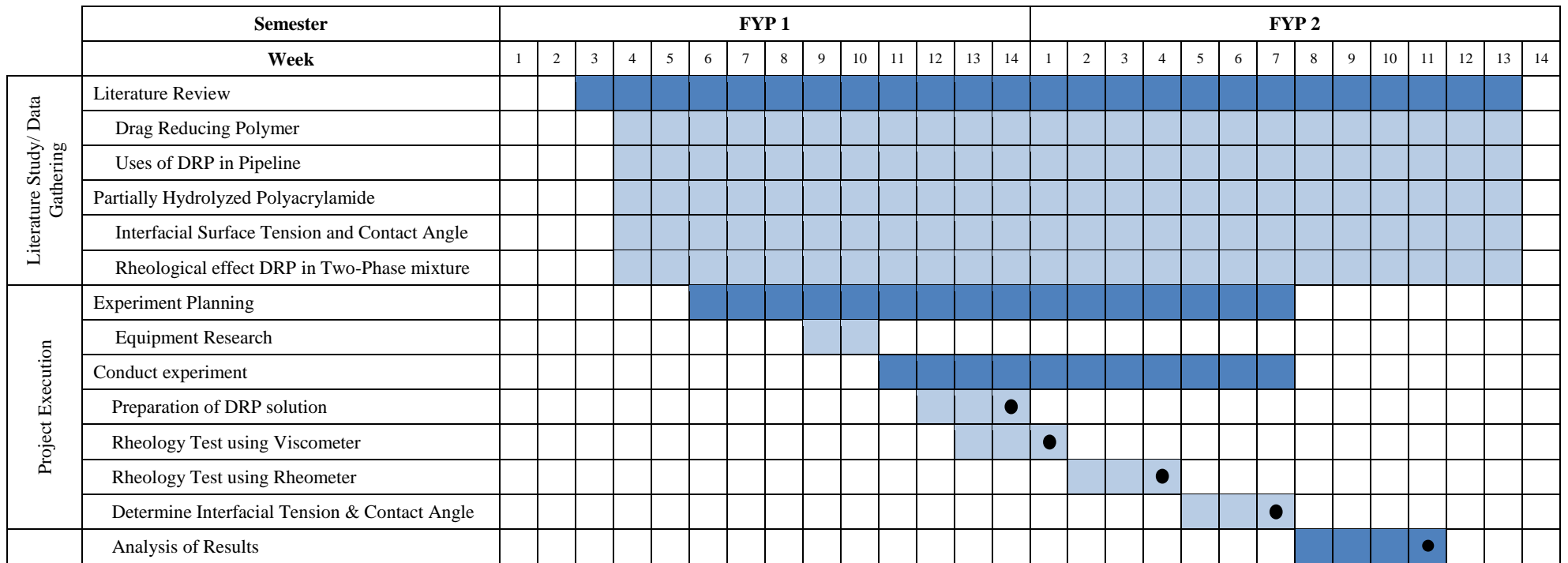


TABLE 3.4 Project Gantt Chart and Milestone

- Process
- Milestone

CHAPTER 4

RESULTS AND DISCUSSION

For this study, the results are analysed qualitatively and quantitatively. For qualitative results, the physical properties of the PHPA solution are analysed before and after it is mixed. Meanwhile, in the qualitative results, the viscosity and density of the PHPA solution are analysed.

4.1 Behaviour of PHPA Solution

The behaviour of the PHPA solution at different concentration shows different characteristic. When the Polyacrylamide powder was first added to the distilled water, the solvent molecules diffuse through the polymer matrix to form a swollen, solvated mass called a gel. After the solution is agitated and left to be hydrated for 24 hours, the gel breaks up and the molecules are dispersed into a homogenous phase. The master solution at 1000 ppm concentration was very viscous and has a gel like consistency. For the diluted solution, the solutions are less viscous compared to the 1000ppm master solution.

4.2 Rheological Properties

Viscometry measurements were carried out using Bohlin C-VOR Rheometer with each of the PHPA samples at a constant shear rate from 0.001s^{-1} to $10,000\text{s}^{-1}$ covering three temperature; 25°C , 40°C and 50°C . The following plots are the results obtained from the viscometry test.

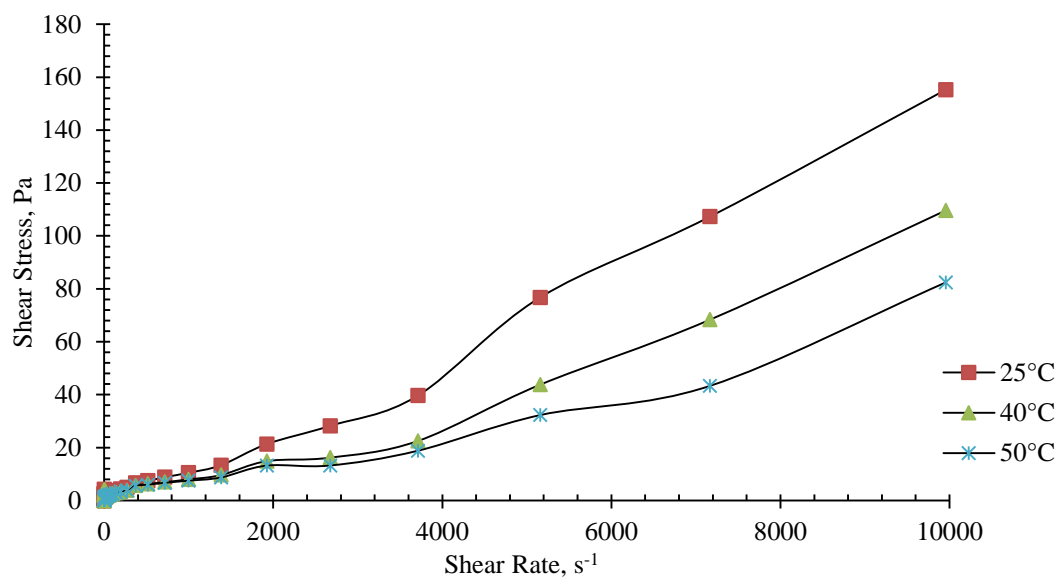


FIGURE 4.1 Shear stress vs shear rate of 1000 ppm PHPA solution at 25 °C, 40 °C and 50 °C

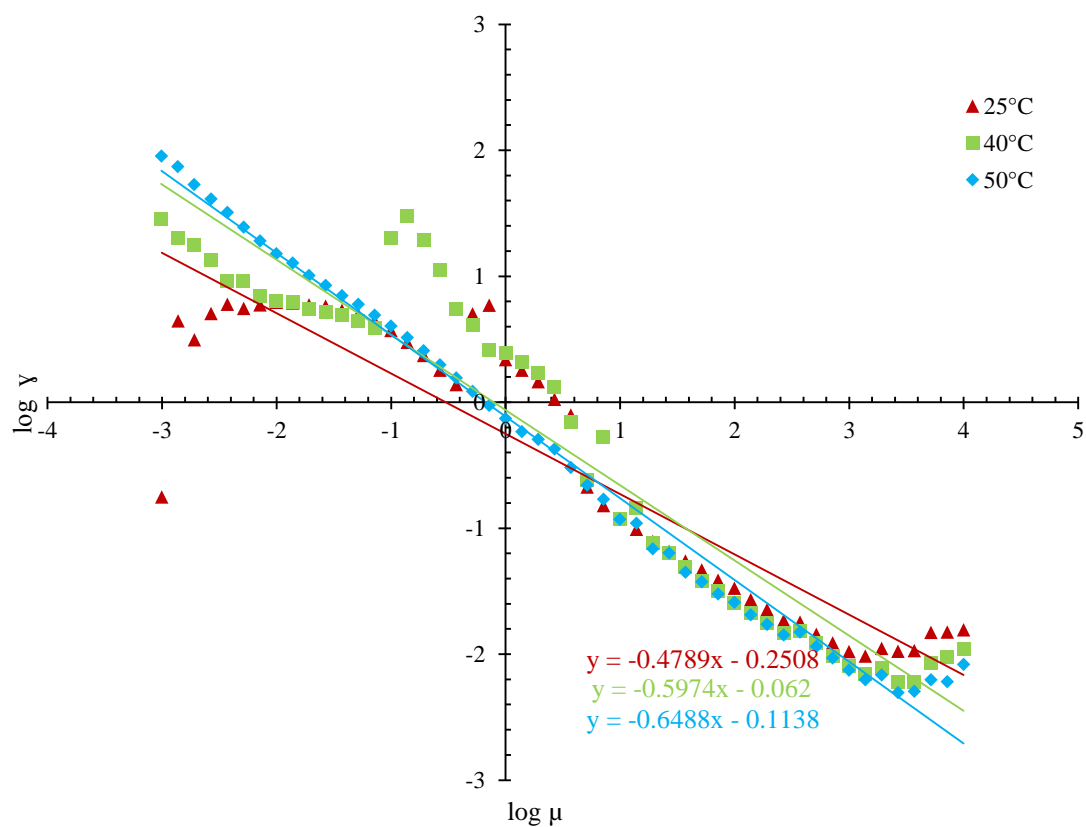


FIGURE 4.2 Log Viscosity vs log shear rate of 1000 ppm of PHPA solution

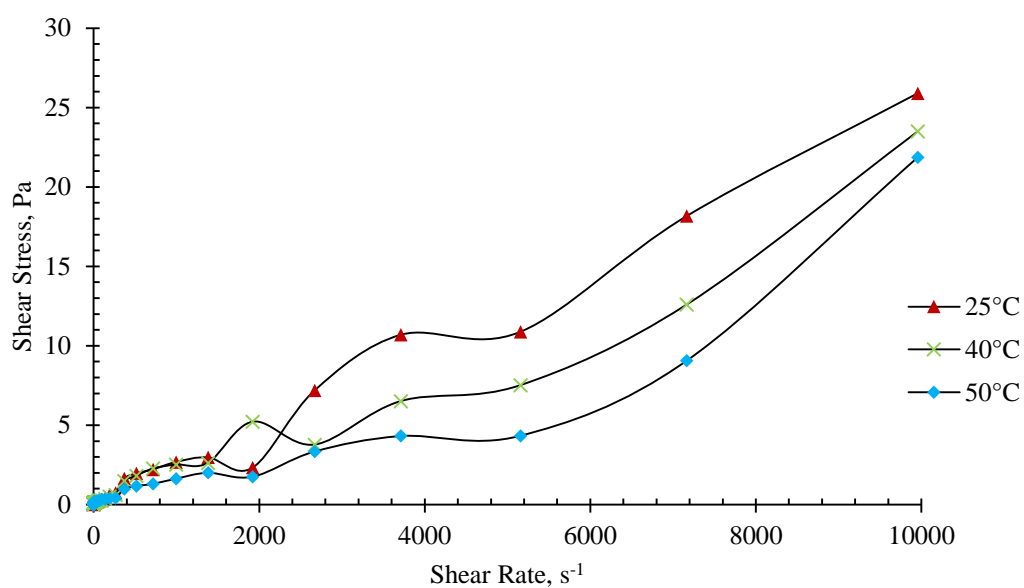


FIGURE 4.3 Shear stress vs shear rate of 10 ppm PHPA solution at 25 °C, 40 °C and 50 °C

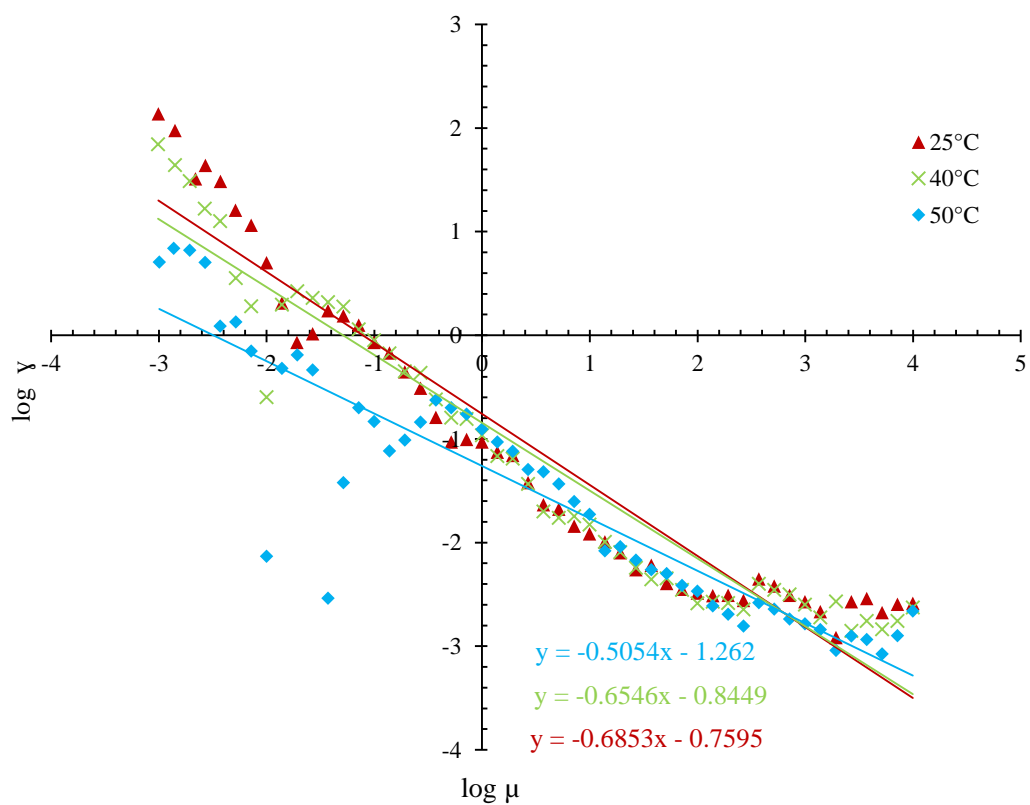


FIGURE 4.4 Log Viscosity vs log shear rate of 10 ppm of PHPA solution

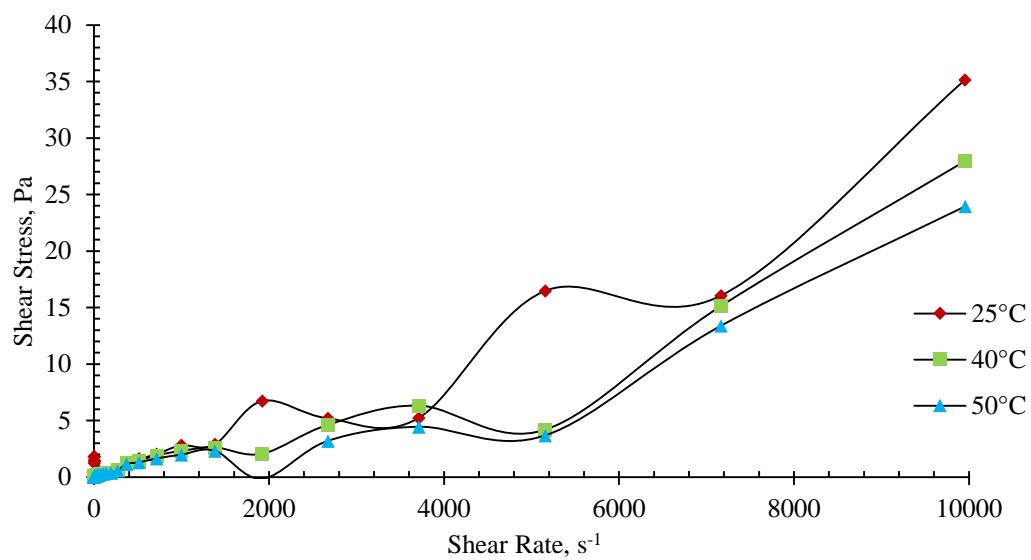


FIGURE 4.5: Shear stress vs shear rate of 20 ppm PHPA solution at 25 °C, 40 °C and 50 °C

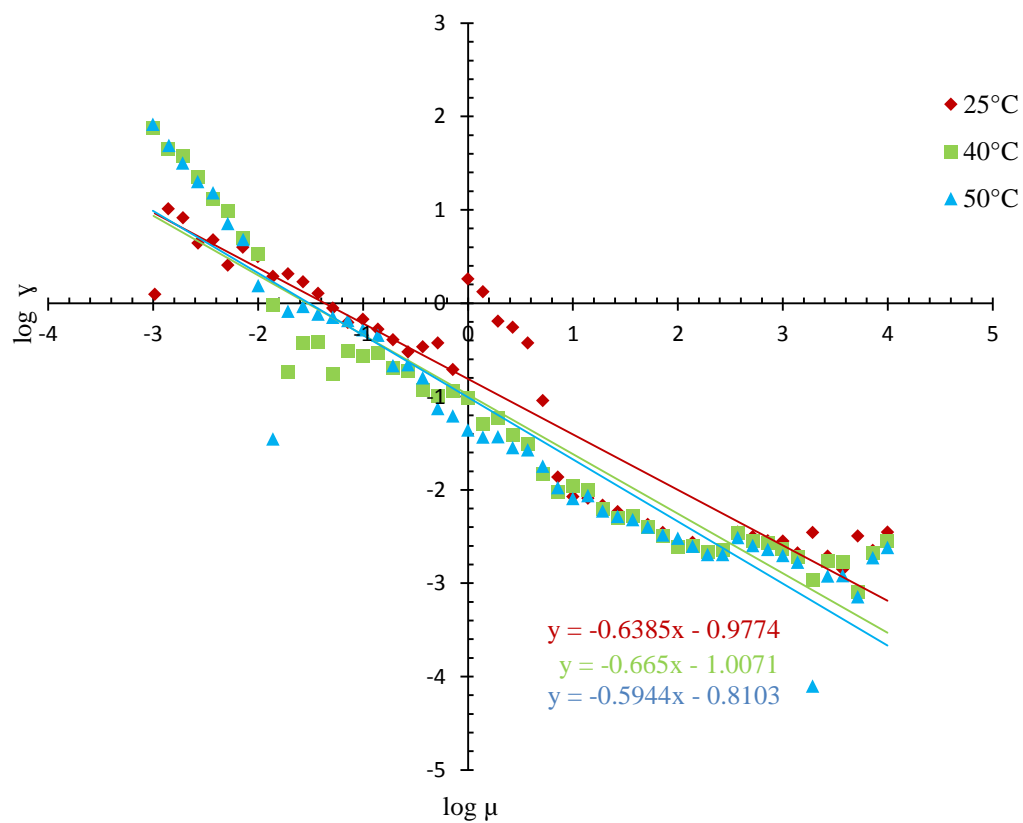


FIGURE 4.6 Log Viscosity vs log shear rate of 20 ppm of PHPA solution

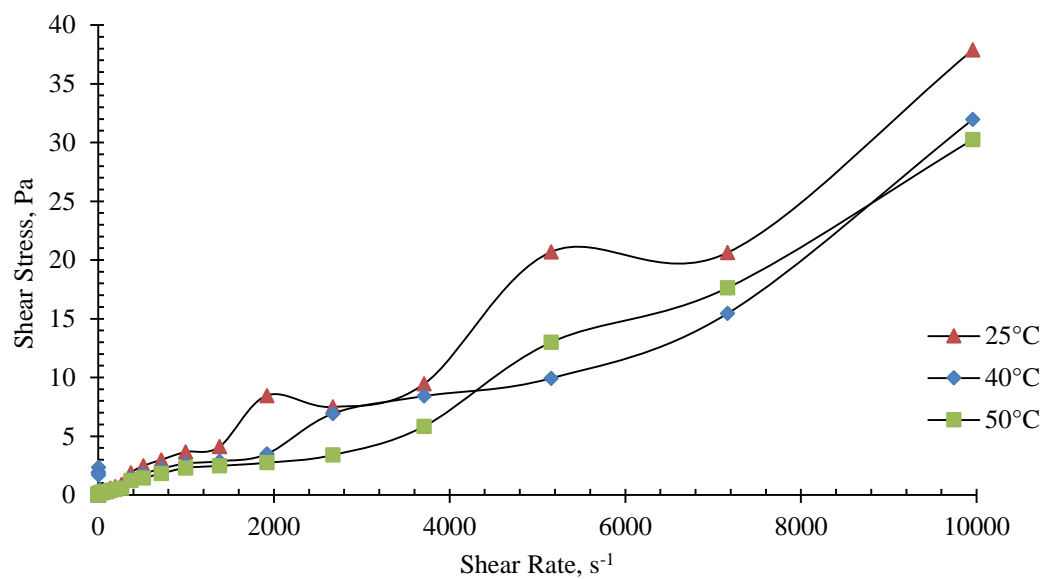


FIGURE 4.7 Shear stress vs shear rate of 50 ppm PHPA solution at 25 °C, 40 °C and 50 °C

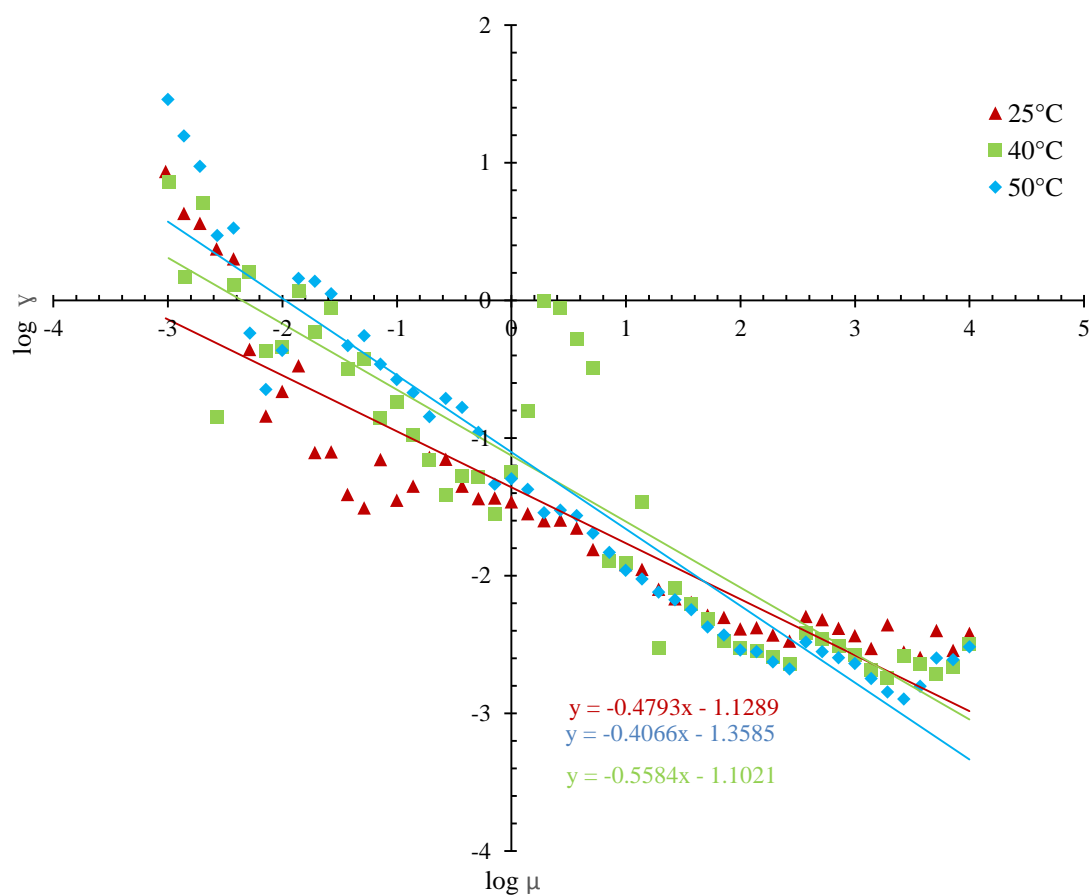


FIGURE 4.8 Log Viscosity vs log shear rate of 50 ppm of PHPA solution

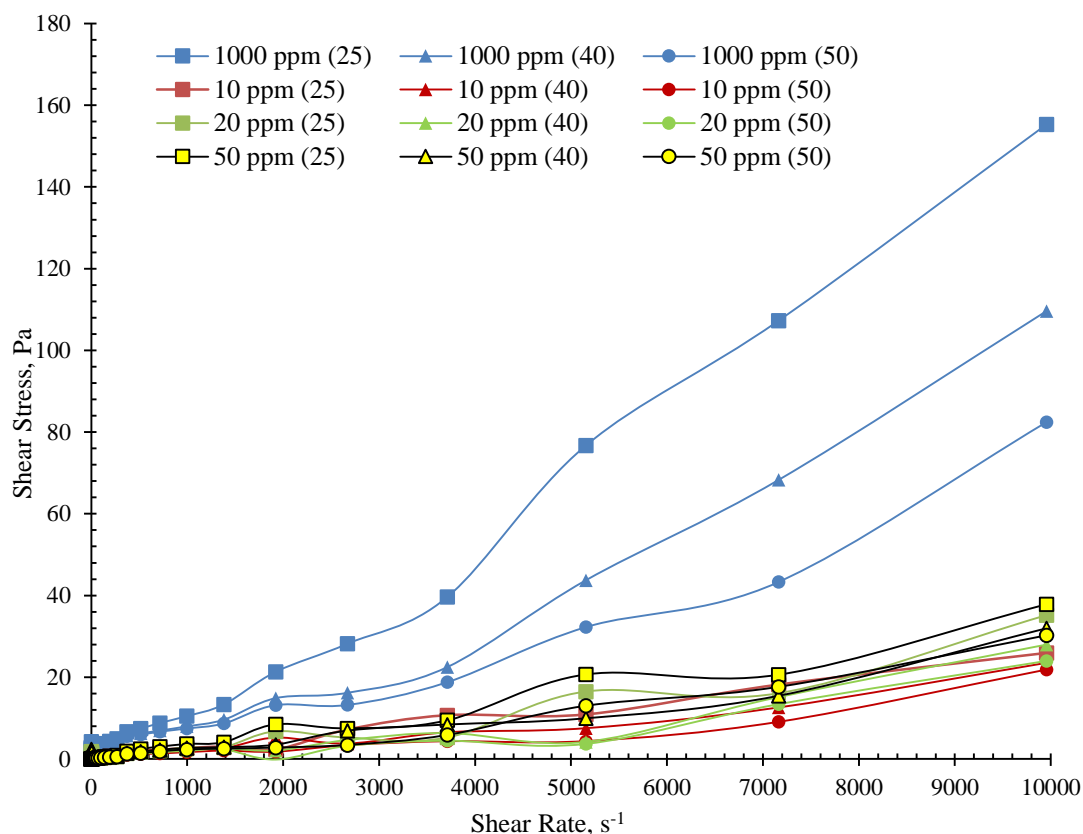


FIGURE 4.9 Shear Stress vs Shear rate at for all PHPA concentration at each temperature

Figure 4-9 shows the effect of increasing shear rates on the solutions' shear stress. As shown in figure, solutions with higher concentration have higher viscosity due to the amount of solute in the solution increases. It is worth noting that the master polymer solution flow at room temperature was difficult compared to the diluted version. Higher concentration of polymer solution has a longer chain of polymer and more cross linked chain due to the hydration period. More solute in the solution causes the bond to be stronger as it is linked together. Another explanation is that polymers are made of coiled chains. When polymers are dissolved into a solution, the charged areas on the chain repel each other and force the chain to uncoil. This causes the viscosity of the solution increases.

Along with shear rate, temperature is also influencing the shear rate. In Figure 4-1,4-3,4-5 and 4-7,the shear stress for all the PHPA solution is the highest at low temperature which is at 25°C compared to when the solution is at 40°C and 50°C.

Similar observation were reported by Al-Shammari, Al-Fariss, Al-Sewailm, and Elleithy (2011) and Gao (2013). According to Al-Shammari et al. (2011), the molecular movement of the polymer is easier at higher temperature due to the increase in polymer solubility. The higher the temperature, the lower the solutions' viscosity. Consequently, decreasing temperature causes an increase in viscosity. The dependence of viscosity, η on temperature, T could be described by Arrhenius equation as shown in Eq. (10).

$$\eta = k_1 \exp\left(\frac{E_0}{RT}\right) \quad (10)$$

Where k_1 is constant, E_0 is activation of energy and R is a gas constant.

From the results, the PHPA solutions investigated here behaved in a non-Newtonian manner as indicated by the reduction of the solution viscosity as the shear rate increases. Under no shear condition, the polymer coil is roughly in spherical in shape. As the polymer solution begins to flow, the flexible polymer coil reacts and the coil deforms as it becomes elongated and aligned to the direction of flow. The shear-thinning viscosity reduction behavior results from the water-soluble polymers becoming uncoiled and untangled when they are aligned and elongated in the fluid-flow shear field under sufficiently high shear-rate conditions. The distorted coil hinders the solution's flow less than the original spherical coil did, and the solution's observed a drop in viscosity as the polymers become less effective viscosity enhancing agents. At high stress, the coils are distorted at maximum and offer low resistance to flow.

An alternative justification of shear thinning is based on assumption of macromolecular cross-linking. When the shear force is applied, it breaks the hydrogen bonds and allows the polymer strands to flow more easily past each other. An average number of cross-links in the shear flow decreases when shear rate is increased, which thus leads to a decreased in apparent viscosity.

A useful form of expressing the flow behavior is the Power Law relationship of the Ostwald de Waele model (Gao, 2013). If the power law parameters (k and n) can be determined, then the polymer solution viscosity is easily calculated under any shear rate.

$$\eta = k\dot{\gamma}^{n-1} \quad (11)$$

$$\log \eta = \log k + (n-1) \log \dot{\gamma} \quad (12)$$

According to Gao (2013), polymer solution is a non-Newtonian fluid that follows the power law equation, where $\dot{\gamma}$ is the shear rate (1/s), τ is the shear stress, η is the shear viscosity, n and k are constants, known as the non-Newtonian index and the consistency index, respectively. By plotting $\log \eta$ with $\log \dot{\gamma}$ as shown in the Figure 4.2, 4.4, 4.6 and 4.8, the value of n and k was calculated.

TABLE 4.1 Table of n values for PHPA at different concentration

PHPA Solution Concentration, ppm	Temperature, °C	n	k
1000	25	0.5211	-0.2508
	40	0.4026	-0.0620
	50	0.3512	-0.1138
10	25	0.3147	-0.7595
	40	0.3454	-0.8449
	50	0.4946	-1.2620
20	25	0.4056	-0.8103
	40	0.3615	-0.9774
	50	0.335	-1.0071
50	25	0.5934	-1.3585
	40	0.5207	-1.1289
	50	0.4452	-1.1021

From the Table 4.1, n values of PHPA solutions at concentration 1000 ppm, 10, 20 and 50 ppm is less than 1 which proves that the solutions exhibit a shear thinning properties. Also, the plot of shear stress vs. shear rate is nonlinear through the origin shows that PHPA solution is a non-Newtonian fluid. Figure 4.10 shows the plot on the Non-Newtonian index, n with respect to temperature. In the figure, all of the solutions exhibit a decrease in the n -value except for 10 ppm. The reason behind the trend displayed by 10 ppm is unclear.

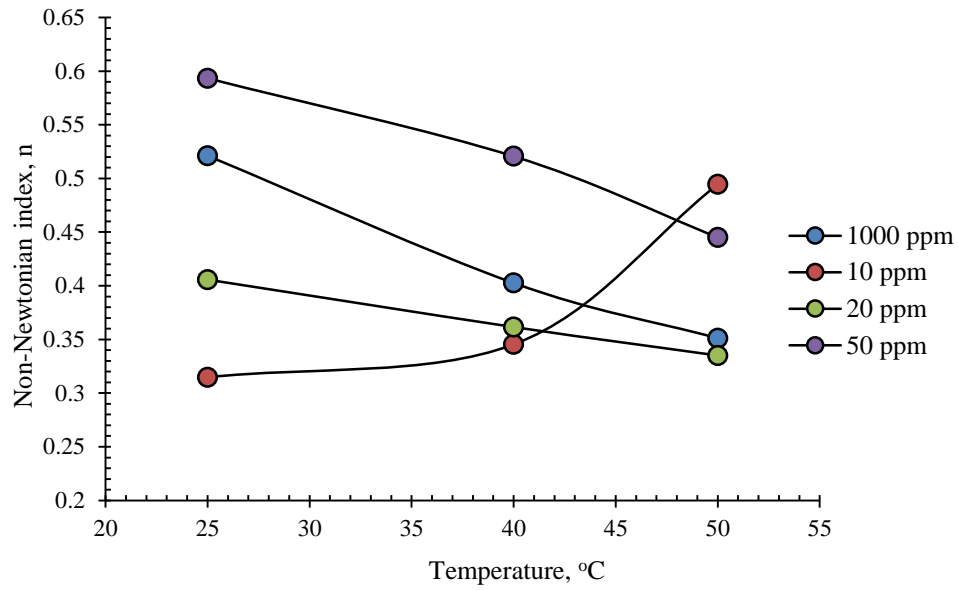


FIGURE 4.10 Non-Newtonian index vs Temperature for all the PHPA solutions

In a flowing two-phase, the viscosity of water is lower than the viscosity of oil, water layer Reynolds number is higher than that for oil layer which then initiates turbulent flow and disturbances waves. With the addition of PDRA, it increases the viscosity of the solution. From the results, it is clear that higher PHPA concentration leads to higher viscosity. It can also be concluded that PHPA viscosity is reduced at higher shear rate and higher temperature.

4.3 Interfacial Tension

This method allows the formation of a drop of one fluid at the tip of a hollow needle which is submerged in the second phase. The drop formation is performed under controlled temperature, with the maximum drop size recorded photographically. Using the drop dimensions on the photographic image, the software used for measuring each of the sample will determine the interfacial tension for the selected fluid. The parameters of the experiment used by the software are listed as below:

Beta	= Shape Factor
R_o	= Radius of curvature at the Drop Apex (mm)
Area	= Drop Surface Area
Volume	= The drop volume
Theta	= Contact Angle at the drop limit (Horizontal) hairline

Height = Total measured height from hairline to apex
Width = Maximum width

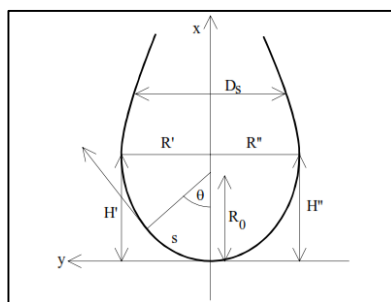


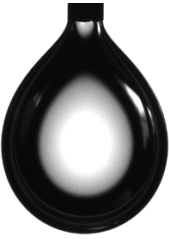
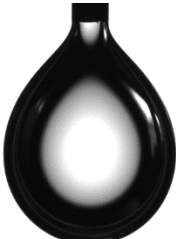
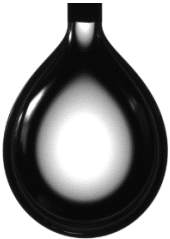

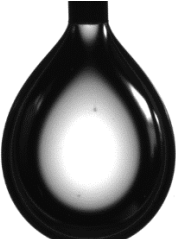

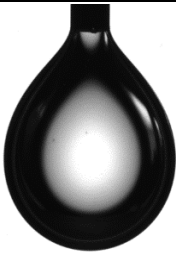
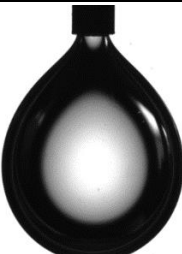
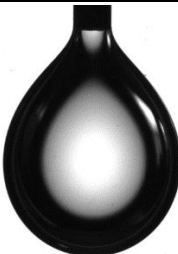
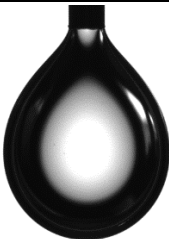
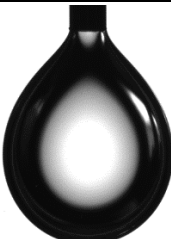

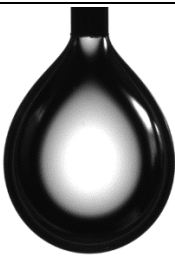
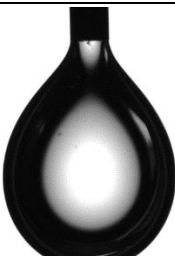
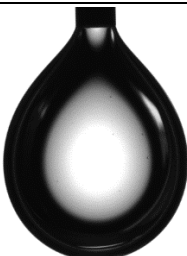
FIGURE 4.10 The pendant drop geometry

The relationship between oil/water interfacial tension and the drop dimensions are as Eq. (9). Table 4.2 shows the results obtained from the Pendant drop experiment.

TABLE 4.2 Data from interfacial tension experiment

Solution	T (°C)	Gamma	Beta	RO	Area	Volume	Theta	Height	Width
Distilled Water	25	19.59	0.21	1.60	38.55	22.22	101.14	4.25	3.32
	40	19.93	0.21	1.62	39.84	23.28	97.59	4.35	3.37
	50	20.80	0.21	1.66	41.44	24.70	97.09	4.44	3.44
PHPA 10ppm	25	18.28	0.19	1.45	30.83	15.97	109.59	3.70	3.00
	40	18.74	0.19	1.48	32.30	17.02	99.81	3.86	3.06
	50	19.94	0.22	1.63	40.09	23.56	101.68	4.34	3.38
PHPA 20ppm	25	19.16	0.22	1.59	38.50	22.16	102.42	4.24	3.32
	40	19.87	0.21	1.60	38.34	22.13	108.92	4.17	3.32
	50	18.92	0.22	1.57	37.30	21.21	107.45	4.13	3.27
PHPA 50ppm	25	17.49	0.22	1.55	36.38	20.39	103.20	4.13	3.22
	40	18.63	0.22	1.58	38.25	21.89	97.46	4.28	3.30
	50	19.31	0.22	1.59	37.95	21.83	109.49	4.15	3.30
PHPA 1000ppm	25	16.89	0.23	1.55	37.02	20.83	98.14	4.24	3.22
	40	16.28	0.24	1.53	35.96	20.04	103.96	4.12	3.18
	50	16.35	0.23	1.52	35.57	19.65	100.99	4.12	3.16

TABLE 4.3 Images of water and PHPA solution in diesel.

	25°C	40°C	50°C
Distilled water in Diesel			
PHPA solution of 1000 ppm in diesel			
PHPA solution of 10 ppm in diesel			
PHPA solution of 20 ppm in diesel			
PHPA solution of 50 ppm in diesel			

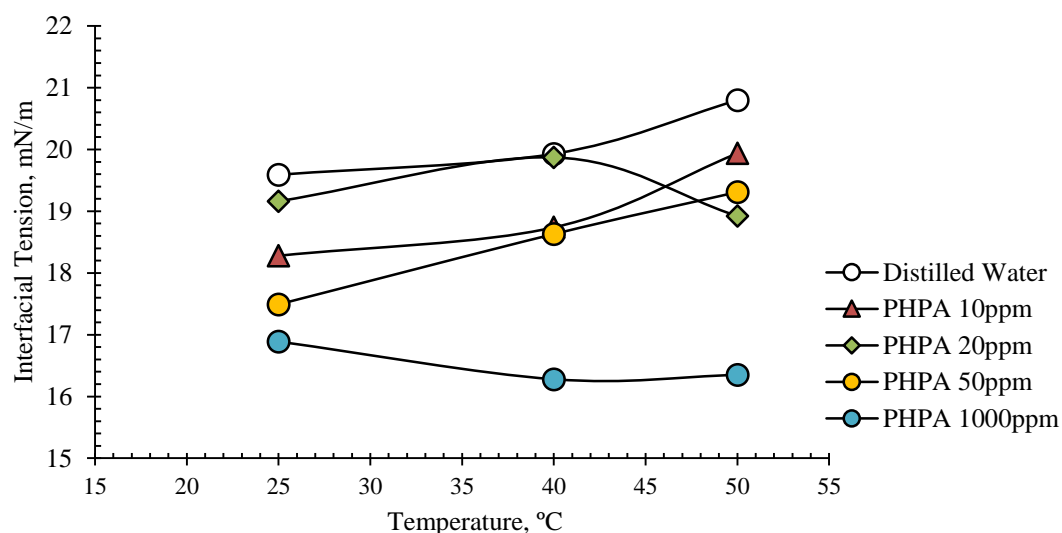


FIGURE 4.11 Interfacial tension as a function of temperature

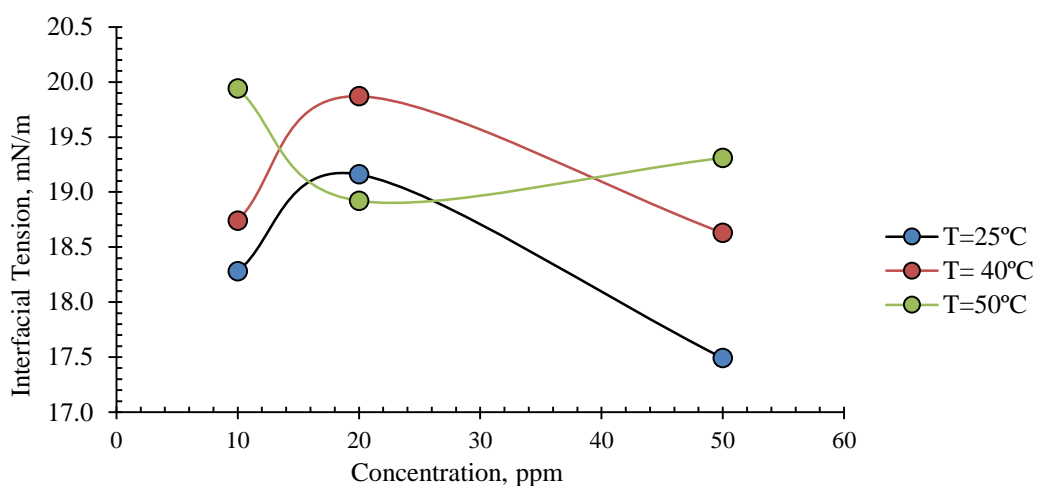


FIGURE 4.12 Interfacial tension as a function of polymer concentration

Figure 4-11 shows the plot of interfacial tension against the temperature. The results shows that the interfacial tension of distilled water and diesel increases as the temperature increases. For all the PHPA solution tested, the interfacial tension between all the solutions with diesel was decreased. The interfacial tension shows a constant trend where it is increasing with temperature except for the PHPA solution with 20ppm and 1000ppm. For 1000 ppm, interfacial tension between the oil-water phase decreases significantly from 19.59 mN/m to 16.89mN/m at 25°C. For PHPA solution at 20 ppm, the interfacial of the oil-water was decreased at 50°C, the lowest compared to 10 ppm and 50 ppm. At 50°C, the possibility of having high energy due to temperature might lower the interfacial tension.

The reduction in the interfacial tension can be explained from the interaction between the molecules of the polymer chain and the diesel. The reduction is due to the decline of the adhesive forces between the molecules of the oil-water interface as the polymer is being introduced to the interface. The polymer molecules tend to be more cohesive towards each other hence reducing the interfacial tension simultaneously making the condition more stable.

Figure 4.12 shows the plot of interfacial tension against the polymer concentration. When comparing in terms of concentration, the results does not show a constant trend for the diluted PHPA solution. However, it can be seen from the graph that the interfacial increases as the temperature increases. Similar trend is observed when the PHPA solution is tested at 25°C and 40°C.

A possible explanation of the relationship between interfacial tension and drag reduction is that the pressure gradient reduction in stratified water layer by PDRA is governed by wall shear stress reduction and interfacial shear reduction between oil and water (Al-Yaari, Al-Sarkhi, & Abu-Sharkh, 2012). After the addition of PDRA, it increases the droplets coalescence rate which can suppress turbulence and a gravity force dominates leading to stratification of water phase which then causes a sharp decrease in turbulence intensity. PDRA promoted the bubbly and dispersed flow into stratified regime, where oil-water is separated via a pronounced interfacial boundary. PDRA minimized most of the interfacial waves and reduced their frequencies which hinders the disturbance waves to form on the oil-water interface (Al-Yaari et al., 2012). From the results obtained, the addition of PHPA decreases the interfacial tension of oil-water system.

The decrease in interfacial tension explained the experimental data obtained by Abdullah, Odjoji, and Angeli (2009) on the effect of polymer concentration on drag reduction. The optimum drag reduction occurs at 20 ppm, whereas low at 10 ppm and 50 ppm. It can be postulated that by reducing the interfacial tension of the oil-water system, the smooth interface hinders interfacial waves and turbulence fluctuations close to the oil-water interface and reduces the pressure losses.

It can be concluded that PHPA solution has a remarkable effect on the interfacial tension between oil and water.

CHAPTER 5

CONCLUSION AND RECOMMENDATION

In this paper, the effect of partially hydrolysed polyacrylamide on the interfacial tension between oil and water was investigated. Three different concentration; 10 ppm, 50 ppm and 70 ppm from a 1000 ppm master solution of PHPA were tested at elevated temperature to study its rheological properties as well as the interfacial tension of the oil-water phase. From the experiment that had been conducted, all solutions that were investigated in this study exhibited “shear thinning”, and is proven as the n value calculated is less than 1. From the results, the PHPA solutions investigated here behaved in a non-Newtonian manner that abide the Power Law. The interfacial tension between the oil-water phases was decreased with the introduction of PDRA due to the reduction of adhesive forces between the molecules of the oil-water interface. By reducing the interfacial tension of the oil-water system, the smooth interface of two-phase flowing system hinders interfacial waves and turbulence fluctuations close to the oil-water interface and assist in reducing the pressure losses. It can be concluded that PHPA solution has a remarkable effect on the interfacial tension between oil and water.

The rheological properties of the PHPA as a PDRA could be used for further study and developed to improve its proficiency in the industry. Further research should be carried out in order to investigate the pattern of the flow of the oil-water phase after PDRA is being introduced in to the pipe. The parameters of the experiment can be broaden for further study for example, testing the interfacial tension at higher temperature. Other than that, different types of polymer as PDRA and oil should be studied in order to investigate the behaviours of the other types of PDRA to the oil-water interphase.

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